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Hybrid membrane cryogenic process for post-combustion Co_2 capture

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Reducing the energy requirement for the capture step is the main challenge in Post-Combustion Carbon Capture and Storage (CCS) technology. In this strategy, the target of 90% capture ratio and 90% CO_2 purity are the main constraints to be satisfied.

Among the different carbon capture processes, the membrane and cryogenic separation processes show a strong sensitivity towards CO_2 content in the flue gas to be treated. More specifically, membrane separation has been shown to offer attractive performance when a moderate purity in the permeate is desired [1-3]. Moreover, the required energy for cryogenic separation is known to be very competitive once a concentrated CO_2 flue gas is treated (approximately > 40%) [4,5]. This suggests that their combination could lead to a minimal overall requirement. This strategy has been already investigated for argon production [6], hydrogen purification [7] and air separation [8] but, to our knowledge, no research has considered this concept for carbon capture framework.

This work aims, through a simulation study, to evaluate the potential of a hybrid process combining membrane and cryogenic separation to achieve efficient Carbon capture.

Figure 1 provides a detailed flow sheet of the process (case of feed compression strategy).

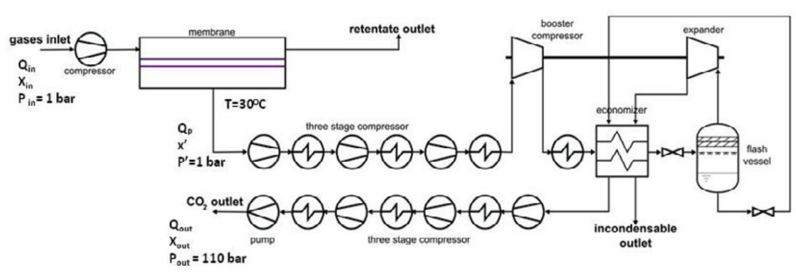

Figure 1: A detailed diagram of the investigated hybrid process.

The inlet feed gas stream, assumed to be a dry binary mixture CO_2/N_2 at temperature of 30°C and under atmospheric pressure, is first separated by a membrane unit to produce a more concentrated CO_2 purity gas stream on the low pressure (permeate) side. A CO_2 capture ratio (R1) of 0.9 is imposed for the simulation of the membrane unit.

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Second, the concentrated CO_2 gas stream is sent to a cryogenic separation unit (based on multi-stage compressors) in order to achieve a CO_2 concentration to the desired purity. The targets imposed in the cryogenic unit are CO_2 purity (x_{out}) higher than 94% and a CO_2 recovery ratio (R_2) above 90%. The intermediate CO_2 mole fraction, noted x', being the key variable to optimise in order to minimise the overall energy requirement of the hybrid process.

The overall energy requirement of the hybrid process, defined as the sum of the energy of the membrane unit E_1 and the cryogenic unit E_2 has been evaluated.

An illustrative example of results is given in figure 2 for sake of hybrid concept validation.

The process is simulated for a power plant flue gas containing 15% CO_2 (x_{in} = 0.15) with a membrane unit using feed compression strategy with an Energy Recovery System (ERS) (using TurboExpander) on the retentate (high pressure side). The membrane selectivity CO_2/N_2 is fixed at 50 (α=50).

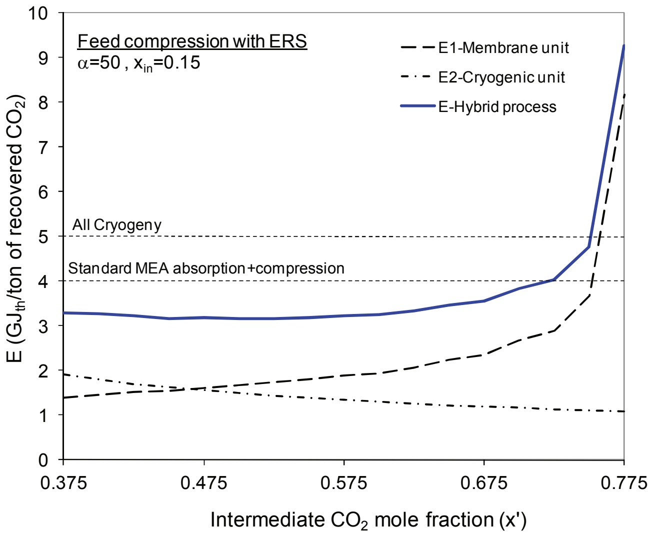

Figure 2: CO_2 specific energy requirement of the hybrid process as a function of intermediate CO_2 mole fraction x' (permeate composition). x_{in}=0.15- α=50 with feed compression + ERS strategy.

The energy requirement of standalone cryogenic process and the reference technology, namely, standard MEA absorption are also reported for sake of comparison. In this figure, it can be seen that the hybrid process significantly decreases the energy requirement compared to the standalone cryogenic option.

Moreover, under optimal conditions (i.e. for x' approximately ranging between 0.5 and 0.6), the energy requirement is competitive with MEA absorption- compression process. This particularly applies for a CO_2 content of 15%, which is the main target of carbon capture studies.

Furthermore and in order to explore the potential interest of improved membrane materials on the energy requirement of the hybrid process, a hypothetical membrane showing a CO_2/N_2 selectivity of 100 and available membrane selectivity of 50 has been simulated for this purpose. Moreover, two compression strategies: (i) Vacuum Pumping, and (ii) Feed Compression with

Energy Recovery System have been investigated and the corresponding overall process performances evaluated and compared.

The results in term of energy requirement and membrane surface area are presented repectively in figure 3 and 4 as a function of the feed mixture CO_2 content x_{in}.

First, it can be seen that the minimum energy requirement decreases, as expected, when CO_2 inlet content x_{in} increases and also when membrane selectivity increases. Second, figure 3 shows that the minimum energy consumption of the hybrid process is very slightly influenced by membrane selectivity (50 or 100). This effect is more important for low mole fraction in the feed, typically x_{in}=0.05.

Moreover, the differences between the compression strategies is more significant with the decrease of x_{in}. Furthermore, the feed compression with turbo expander and permeate vacuum stratgies have generally similar values, with the pemeate vacuum energy being slightly lower.

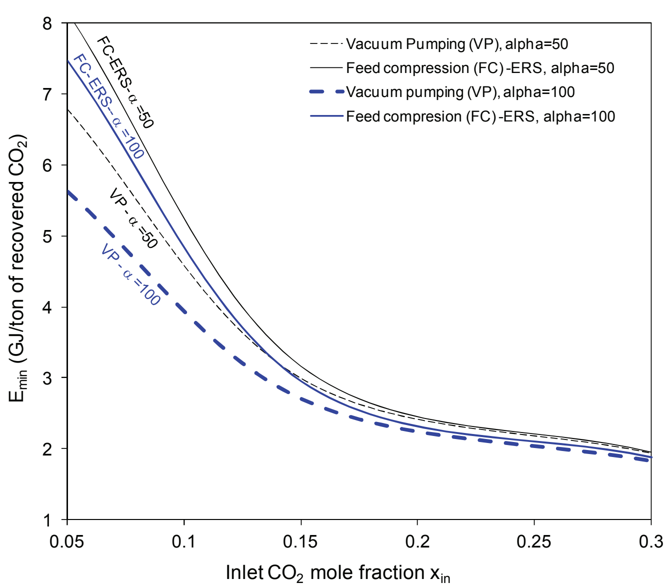

Figure 3: Evolution of E_{min} as a function of CO_2 mole fraction in the feed, different compression strategy and membrane selectivity (α=50 and 100).

In figure 4, one can observe that logically the required membrane surface is very high for vacuum pumping strategy compared to feed compression strategy, a factor of 20 is attained. That suggests that the use of feed compression with Energy Recovery System (ERS) configuration offers the best performances when energy requirement and membrane surface area are both taken into account. Moreover, it can be seen that more selective membrane requires higher membrane surface area, the effect being more important when inlet CO_2 content decreases.

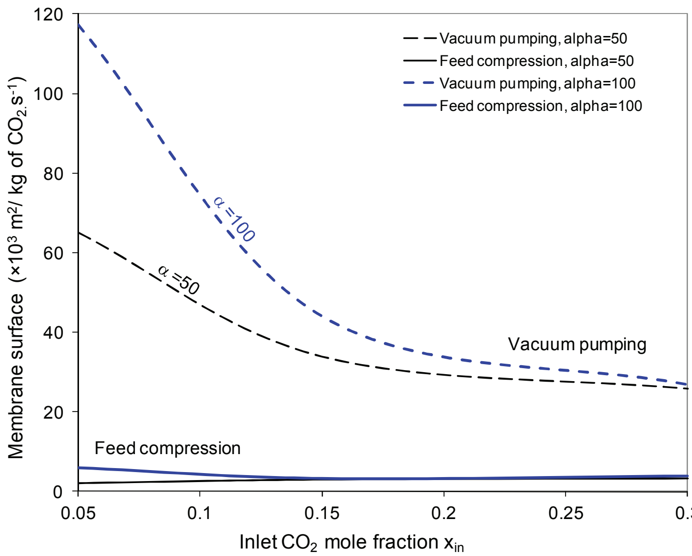

Figure 4: Evolution of membrane surface area as a function of CO_2 mole fraction in the feed. Two compression strategies in the membrane unit, α= 100 (bold line), membrane CO_2 permeance is fixed at 1000 Gas Permeation Unit (GPU).

Finally, figure 5 tries to give a tentative process selection map for post-combustion CO_2 capture.

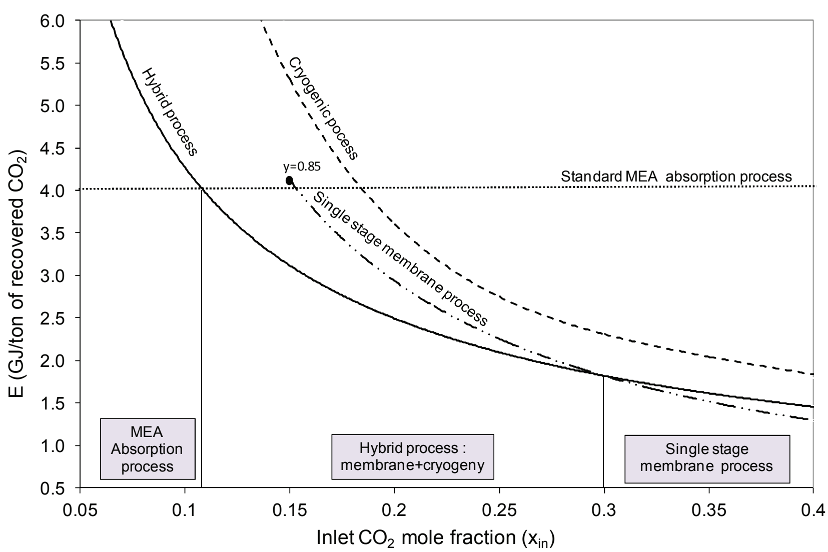

Figure 5: A schematic process selection map for post-combustion CO_2 capture as a function of the inlet CO_2 mole fraction- Membrane selectivity =100 - CO_2 purity : y>=90% and total recovery

ratio : R>=0.85. Framed processes correspond to the more energy efficient process for different x_{in} ranges.

One can remark that the hybrid process effectively improves the energy efficiency compared to a standalone cryogenic process. The percentage energy decrease significantly increases for lower inlet CO_2 mole fractions. For concentrated CO_2 inlet content (i.e. > 30%), a single stage membrane process is the more energy efficient process. One can remark that for very diluted CO_2 concentration (i.e. <15%), a multistage membrane process is needed in order to attain the CO_2 purity (y=0.9) and capture ratio (R=0.9)[9].

Compared to the MEA absorption technology, the hybrid process offers improved energy performances for CO_2 inlet content ranging between 10% and 30%. This corresponds to a large proportion of emission sources, such as coal power plant flue gases (for the lower range) and oxygen enriched air combustion (for the upper range). The hybrid process is however too energy intensive for a CO_2 content lower than 12%, such as natural gas turbine exhaust gases.

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